PTO 07-6023

CC=JP DATE=19940408 KIND=A
PN=06096783

FUEL CELL
[Nenryou denchi]

Makoto Uchida, et al.

UNITED STATES PATENT AND TRADEMARK OFFICE
Washington, D.C. August 2007

Translated by: FLS, Inc.

PUBLICATION COUNTRY	(19):	JP
DOCUMENT NUMBER	(11):	06096783
DOCUMENT KIND	(12):	A [PUBLISHED UNEXAMINED APPLICATION]
PUBLICATION DATE	(43):	19940408
APPLICATION NUMBER	(21):	04246201
APPLICATION DATE	(22):	19920916
INTERNATIONAL CLASSIFICATION	(51):	H01M 8/02
INVENTORS	(72):	UCHIDA, MAKOTO; AOYAMA, HIROKO; EDA, NOBUO
APPLICANT	(71):	MATSUSHITA ELECTRIC INDUSTRY CO., LTD.
TITLE	(54):	FUEL CELL
FOREIGN TITLE	(54A):	NENRYOU DENCHI

[Claims] /2*

[Claim 1] With respect to a fuel cell in which unit cells comprising positive electrodes, electrolyte plates, and negative electrodes are layered via separator sheets with gaskets disposed on the rims of the unit cells, a fuel cell in which each of the gaskets is made up of a closed-bubble sponge sheet.

[Claim 2] With respect to a fuel cell in which unit cells comprising positive electrodes, electrolyte plates, and negative electrodes are layered via separator sheets with gaskets disposed on the rims of the unit cells, a fuel cell in which each of the gasket has an integrated structure in which closed-bubble sponge sheets are adhered to both sides of a metal substrate.

[Claim 3] With respect to a fuel cell in which unit cells comprising positive electrodes, electrolyte plates, and negative electrodes are layered via separator sheets with gaskets disposed on the rims of the unit cells, a fuel cell in which each of the gasket has an integrated structure with rubber sheets attached to both sides of a metal substrate and in which the parts surrounding the sealing parts of the gasket are subjected to a mountain-shaped embossing process.

[Claim 4] With respect to a fuel cell in which unit cells, each of which comprises an ion-exchange film composed of a solid polymer and a positive electrode and negative electrode equipped with electrode catalytic layers on both sides that are in contact with the ion-exchange film, a fuel cell made up of the gaskets of any one of Claim 1, Claim

* Numbers in the margin indicate pagination in the foreign text.

2, or Claim 3.

[Detailed Description of the Invention]

[0001]

[Field of the Invention] The present invention relates to a fuel cell which employs a reducing agent, such as pure hydrogen or modified hydrogen from methanol or a fossil fuel, as fuel and which employs air or oxygen as an oxidizing agent, specifically to gaskets for polymer electrolyte fuel cells.

[0002]

[Related Art] For example, it is known that when a cation-exchange membrane, which is a proton conductor, is utilized as the solid polymer electrolyte in a polymer electrolyte fuel cell and when hydrogen and oxygen are introduced as fuel and oxidizing agent, respectively, the following reactions occur.

[0003]

(Chemical Formula 1) Negative electrode $H_2 \rightarrow 2H^+ + 2e^-$

(Chemical Formula 2) Positive electrode $1/2O_2 + 2H^+ + 2e^- \rightarrow H_2O$

At the negative electrode, hydrogen becomes dissociated into protons and electrons. Protons move through the cation-exchange membrane toward the positive electrode, and electrons move through conductive separator sheets, serially layered cells, and further through an external circuit, and reach the positive electrode. At this time, electricity is generated. Incidentally, the protons that traveled through the cation-exchange membrane, electrons that traveled through the external circuit, and oxygen introduced from the exterior react with one another and produce water

at the positive electrode. Since this reaction accompanies heat generation, overall, electricity, water, and heat are produced from hydrogen and oxygen.

[0004] A polymer electrolyte fuel cell is greatly different from other types of fuel cells in that the electrolyte is made up of an ion-exchange membrane that is a solid polymer. For this ion-exchange membrane, a perfluorocarbon sulfonic acid polymer membrane (made by DuPont Co., USA, trade name: Nafion). In order for this membrane to demonstrate sufficient proton conductivity, the membrane needs to be fully hydrated. As an example of the method employed to hydrate the ion-exchange membrane, the ion-exchange membrane is prevented from being dried by introducing water vapor into the cell by passing the reaction gas through a humidifier as indicated in J. Electrochem. Soc., 135(1988)2209. Moreover, as an example of the method for sealing the individual cells, the areas of the ion-exchange membranes are made to be larger than the areas of the electrodes and the rims of the ion-exchange membranes not in contact with the electrodes are sandwiched by top and bottom gaskets as mentioned in J. Power Sources, 29(1990)367. As the material of the gaskets, glass cloth or fluorocarbon rubber obtained by coating polytetrafluoroethylene (made by DuPont Co., USA, trade name: Teflon) is utilized. According to U.S. Patent No.4,826,741, silicon rubber or fluorocarbon rubber is utilized. During this formation, the gaskets need to insulate the neighboring separator sheets and seal them from the gas while absorbing the thickness of the ion-exchange membranes which are between about 150 and 200μm. Consequently, these require microscopic processes in order to collapse

the gaskets by increasing the cell tightening force and to reduce the thickness of the portions of the ion-exchange membranes that touch the gaskets as much as the thickness of the membranes.

[0005]

[Problems that the Invention is to Solve] However, according to the above conventional method, the total thickness of the ion-exchange membranes to be absorbed becomes integrated and increases as the number of cell layers increases and makes the absorption impossible, or a very large tightening pressure is required and the other components of the housing such as the end plates and bolt nuts become large in order to ensure the strength. There is also a shortcoming in that sufficient gas-sealing properties cannot be ensured due to the variations in the thicknesses of the gaskets, ion-exchange membranes, and separator sheets. Furthermore, since the thicknesses of the ion-exchange membranes are altered as their water-content ratios change, there is a risk in that the sealing properties of the conventional gasket material which were initially ensured decrease during operation due to its high stress-relaxation properties.

[0006] In order to overcome the above conventional problem, the present invention is aimed at providing a lighter and more economical fuel cell, specifically a polymer electrolyte fuel cell, by utilizing gaskets that demonstrate high sealing properties in response to a low tightening pressure.

[0007]

[Means for Solving the Problems] With respect to a fuel cell in which

unit cells comprising positive electrodes, electrolyte plates, and negative electrodes are layered via separator sheets with gaskets disposed on the rims of the unit cells, the invention, in order to achieve the above aim, has a structure in which each of the gaskets is made up of a closed-bubble sponge sheet, in which each of the gaskets has an integrated structure in which closed-bubble sponge sheets are adhered to both sides of a metal substrate, or in which each of the gaskets has an /3
integrated structure with rubber sheets attached to both sides of a metal substrate and in which the parts surrounding the sealing parts of the gasket are subjected to a mountain-shaped embossing process.

[0008]

[Operation of the Invention] According to the first structure, the closed-bubble sponge sheets absorb the thicknesses of the ion-exchange membranes by means of the compression of the bubbles. Moreover, since the individual closed bubbles also become compressed in response to sectional unevenness, the waviness and roughness of the separator sheets can also be absorbed. Moreover, since the airtight bubbles are compressed, the stress relaxation is small. According to the second structure, the sponge sheets do not become dislocated to the exterior because of the adhesive force between the substrate and sheets even when high-pressure gas is used inside. Since a metal sheet is used as the substrate, highly accurate processing is possible. For example, if an aluminum sheet or the like is used as the substrate, punching by means of a Thompson mold will be easy to perform. According to the third structure, the mountain-shaped embossing of the parts surrounding the sealing part

enables linear sealing that is similar to that of an O-ring.

[0009]

[Embodiment of the Invention] In the following, an embodiment of the invention will be explained by referring to the accompanying drawings.

[0010] Figure 5 is an external view of the layered structure of a general polymer electrolyte fuel cell. Separator sheets 2 composed of a conductive raw material, such as glassy carbon, and insulating gaskets 1 are layered alternately, and current collectors 3 made of copper are firmly attached to the outermost separator sheets. This layered body is sandwiched by stainless-steel end plates 5 via insulating sheets 4, and the end plates are tightened together by means of bolts and nuts. Needless to say, the materials of the individual parts are not limited to the above materials as long as such conditions as conductivity, insulating properties, heat-insulating properties, and gas permeability do not have adverse effects on the cell performance.

[0011] Figure 6 shows a cross-sectional drawing of the internal cell of a general stacked cell. Electrodes 12 are connected to either side of an ion-exchange membrane 11 at the center, and separator sheets 2 having grooves are disposed over and below the connected body. The area of the ion-exchange membrane is larger than that of the electrodes, and its surrounding parts are sandwiched by gaskets to seal each cell and to insulate the separator sheets from each other. As illustrated in the figure, if forming gas passages 13 inside the layered body as necessary (i.e. internal manifold type), the gaskets seal these gas passages, as well. The separator sheets with grooves can have various structures in which,

for example, groove-equipped porous sheets are fitted into the groove areas or in which meshes are utilized, and the above structure does not limit the invention.

[0012] (Embodiment 1) Figure 1 shows a cross-sectional drawing of the cell of embodiment 1 of the invention. As the gasket 21, a 1.0mm-thick silicon-made closed-bubble sponge sheet made by Kureha Gomu Kogyo was utilized. In the case of the gasket of the invention, the closed bubble 22 of the portion that was in contact with the ion-exchange membrane became compressed even more than the part sandwiched by the separator sheets 2. As a result, the sealing of the separator sheets as well as the sealing of the ion-exchange film and separators could be accomplished while absorbing the thickness of the ion-exchange film 11. As for the tightening pressure, the gaskets of the invention only required a sealing pressure of 3kg/cm^2 or more while conventional fluorocarbon rubber not containing bubbles requires 10kg/cm^2.

[0013] (Embodiment 2) Figure 2 shows a cross-sectional view of the cell of embodiment 2 of the invention. As the gasket 31, a foam rubber sheet (trade name: Metafoam) made by Nichias Corp. was utilized. This gasket was obtained by adhering closed-bubble sponge rubber 33 composed of butadiene and acrylonitrile rubber to both sides of a 0.25mm-thick aluminum sheet 32, which made up the substrate, and the overall thickness was made to be 1.5mm. Because of the similar sealing effect as that of Embodiment 1, only 2kg/cm^2 or more was sufficient as the sealing pressure. Furthermore, while a conventional fluorocarbon rubber gasket or the sponge sheet of embodiment 1 became dislocated to the exterior and then torn

away when the internal pressures of the cells and gas passages become high, the gasket of Embodiment 2 prevented the dislocation, and therefore the tearing, of the rubber layers because of the adhesive force between the aluminum sheet and the butadiene and acrylonitrile rubber layers.

[0014] (Embodiment 3) Figure 3 and Figure 4 of Embodiment 3 of this invention show a cross-sectional perspective drawing of the gasket and a cross-sectional drawing of the cell of the invention, respectively. As the gasket, a metal/rubber composite gasket (trade name: Metacoat) made by Nichias Corp. was utilized. This gasket 41 was obtained by adhering nitrile rubber 43 to both sides of a 0.25mm-thick iron sheet 42, which made up the substrate, and the overall thickness was made to be 0.38mm. The parts surrounding the electrodes and the parts surrounding the gas feeding holes were subjected to embossing 44. The heights of the embossing of the parts surrounding the electrodes were made to be slightly lower in order to sandwich an ion-exchange membrane. This mountain-shaped embossing of the parts surrounding the sealing part enabled linear sealing similar to that of an O-ring, and $6kg/cm^2$ or more was sufficient as the sealing pressure.

[0015] Although the above materials were utilized for the gaskets in the present embodiments, various elastic materials can be utilized for these polymer electrolyte fuel cells which operate at temperatures no more than 120°C, and they require no special chemical resistance since they do not employ a corrosive electrolytic solution. Therefore, any material can be selected as long as heat resistance can be ensured at 120°C, and the invention is not limited to the materials of the embodiments.

[0016] Moreover, although the method in which a single gasket is used to seal the ion-exchange membrane from one direction was illustrated in the embodiments, similar effects could be achieved by sandwiching the ion-exchange membrane by using two gaskets instead. Moreover, although polymer electrolyte fuel cells were described as examples in the embodiments, similar effects could be demonstrated for phosphoric-acid fuel cells, alkaline fuel cells, etc.

[0017]

[Effects of the Invention] As described in the above, the invention is the structure of a fuel cell in which the gasket is made up of a closed-bubble sponge sheet, in which the gasket has an integrated structure in which closed-bubble sponge sheets are adhered to both sides of a /4 metal substrate, or in which the gasket has an integrated structure with rubber sheets attached to both sides of a metal substrate and in which the parts surrounding the sealing parts of the gasket are subjected to a mountain-shaped embossing process. Therefore, according to the first structure, the closed-bubble sponge sheets absorb the thickness of the ion-exchange membrane and unevenness of the separator sheets by means of the compression of the bubbles. Therefore, an excellent sealing performance can be realized with a small tightening pressure. According to the second structure, since the closed-bubble sponge sheets are attached to the substrate made of a metal, the adhesive force prevents the sponge sheets from being dislocated to the exterior even when high-pressure gas is utilized inside. Also, since a metal sheet is used as the substrate, highly accurate processing is possible. According to the third structure,

the mountain-shaped embossing of the parts surrounding the sealing part enables linear sealing that is similar to that of an O-ring.

[0018] The above effects allow for a significant reduction in the tightening pressure, which makes it possible to reduce the strengths of the end plates and/or separators. For example, stainless steel conventionally utilized for the end plates can be replaced by materials such as engineered plastic, and small, light-weight, and highly economical fuel cells can be realized as a result.

[Brief Description of the Drawings]

[Figure 1] A cross-sectional drawing of the cell of Embodiment 1 of the invention.

[Figure 2] A cross-sectional drawing of the cell of Embodiment 2 of the invention.

[Figure 3] A cross-sectional perspective drawing of the gasket of Embodiment 3 of the invention.

[Figure 4] A cross-sectional drawing of the cell of Embodiment 3 of the invention.

[Figure 5] An external drawing of a common polymer electrolyte fuel cell.

[Figure 6] A cross-sectional drawing of a common cell.

[Description of the Reference Numerals]

1 = gasket

2 = separator sheet

3 = current collector

4 = insulating sheet

5 = end plate

6 = hydrogen inlet

7 = hydrogen outlet

8 = oxygen inlet

9 = oxygen outlet

10 = water discharge drain

11 = ion-exchange membrane

12 = electrode

13 = gas passage

21 = gasket of embodiment 1

31 = gasket of embodiment 2

32 = aluminum sheet

33 = sponge rubber layer

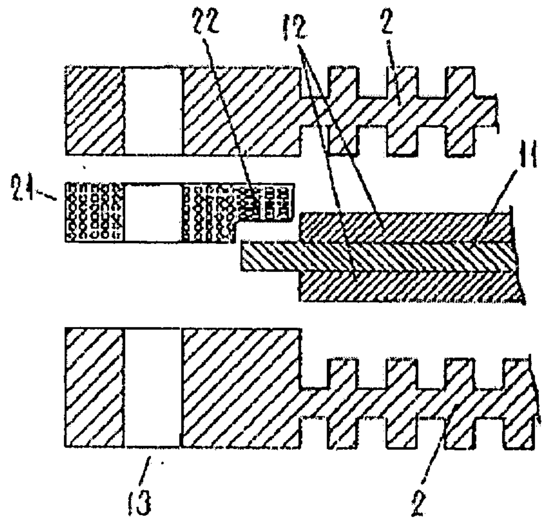
41 = gasket of embodiment 3

42 = iron sheet

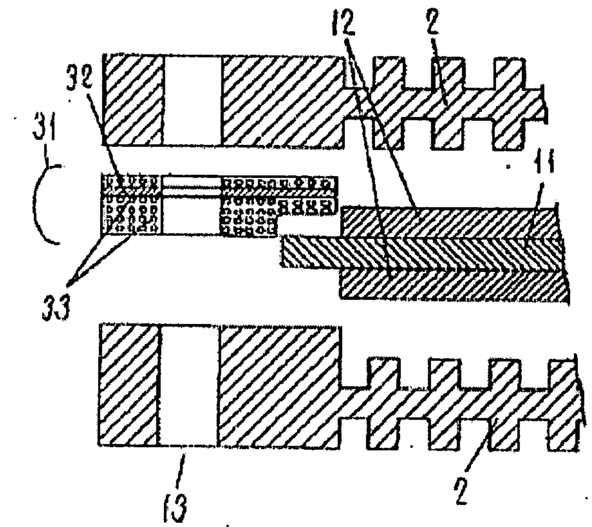
43 = rubber layer

44 = embossing

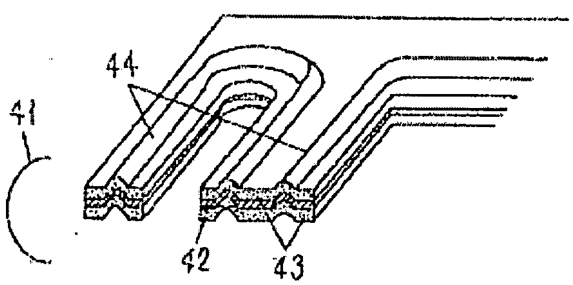
[Figure 1]



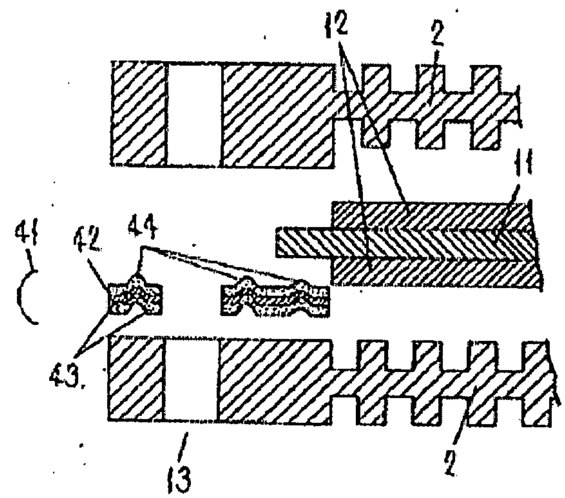
[Figure 2]



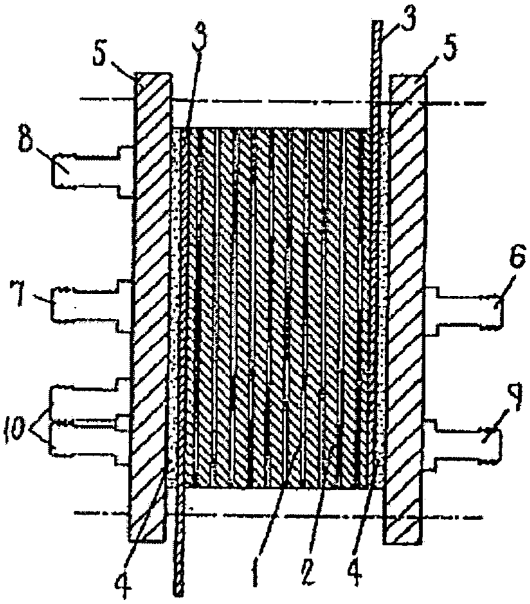
[Figure 3]



[Figure 4]



[Figure 5]



[Figure 6]